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2019-12

Asmala , E , Carstensen , J & Räike , A 2019 , ' Multiple anthropogenic drivers behind upward trends in organic carbon concentrations in boreal rivers ' , Environmental Research Letters , vol. 14 , no. 12 , 124018 . <https://doi.org/10.1088/1748-9326/ab4fa9>

<http://hdl.handle.net/10138/311285>

<https://doi.org/10.1088/1748-9326/ab4fa9>

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To cite this article: Eero Asmala *et al* 2019 *Environ. Res. Lett.* **14** 124018

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Environmental Research Letters



LETTER

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OPEN ACCESS

RECEIVED
4 July 2019REVISED
18 October 2019ACCEPTED FOR PUBLICATION
21 October 2019PUBLISHED
27 November 2019

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Increases of riverine organic carbon concentrations have been observed across the northern hemisphere over the past few decades. These increases are the result of multiple environmental drivers, but the relative importance of the drivers is still unclear. We analyzed a dataset of >10 000 observations of riverine total organic carbon (TOC) concentrations and associated water chemistry and hydrological observations from 1993 to 2017. The observations span a ~600 km north–south gradient from 30 individual river systems in Finland. Our data show significantly increasing TOC concentrations in 25 out of 30 systems, with an average increase from 12.0 to 15.1 mg l⁻¹. The observed increase in riverine TOC concentrations led to an increase of 0.28 Mt in annual TOC load to the Baltic Sea from 1993 level to 2017 level. We analyzed the role of three putative environmental drivers of the observed TOC trends. Multiple regression analysis revealed that the most common driver was discharge, which alone explained TOC increases in 13 rivers, whereas pH and temperature were less important drivers (sole predictor in one and zero rivers, respectively). Different permutations of these three drivers were also found to be significant; the combination of discharge and pH being the most common (4 rivers). Land use was not in general linked with trends in TOC, except for the proportion of ditched land in the catchment, which was significantly correlated with increases in TOC concentration. Land use showed significant relationships with trends in discharge and pH. We also found that catchment characteristics are regulating the extent of these regional or global environmental changes causing the upward trends of riverine organic carbon.

Introduction

Climate change and land use change are profoundly affecting the global carbon cycle and causing drastic shifts in functioning of ecosystems worldwide (Regnier *et al* 2013). Boreal systems are expected to be strongly influenced, as projected increases in temperatures are particularly high in these areas. Furthermore, precipitation is also expected to increase in boreal areas, enhancing freshwater transport from catchments to the coastal sea. Since soils in boreal areas contain one third of the global soil organic carbon (OC) pool (Settele *et al* 2015), even minor changes in the balance of carbon sequestering and exporting in these catchments have potentially large implications

on a global scale. Long-term increases in riverine OC concentrations have been reported throughout the northern hemisphere (Filella and Rodríguez-Murillo 2014). As rivers are important conduits linking terrestrial and oceanic ecosystems by transporting soil organic carbon from land to sea, changes in hydrology or water chemistry will have an effect on the coastal environment as well. The observed increases in riverine fluxes of OC contribute to coastal eutrophication, potentially exacerbating its adverse effects on coastal ecosystems, such as hypoxia (Conley *et al* 2011) and loss of biodiversity (Villnäs and Norkko 2011). Further, increased OC concentrations are also indicative of the potential loss of soil organic carbon to aquatic systems (Tranvik and Jansson 2002).

Most likely, the observed OC trends cannot be explained by one single variable alone, as the increasing OC results from complex interactions of different environmental drivers (Evans *et al* 2006, Clark *et al* 2010, Couture *et al* 2012). The effect of a single driver changes from system to another, suggesting a complex interplay of different environmental factors determining the response of aquatic OC concentration to these drivers. OC concentrations and OC export to the Baltic Sea from Finnish rivers have increased in recent decades (Räike *et al* 2012, 2016), and at the same time the OC content in Finnish cropland soils have decreased (Heikkinen *et al* 2013). However, the question about the role of different environmental drivers behind the increasing OC concentrations has remained open.

Climate change and the associated warming and increasing precipitation have been attributed as the main cause behind increased OC concentrations in a number of systems (Hongve *et al* 2004, Keller *et al* 2008, Tank *et al* 2016). As water balance of the catchments is a major driver of carbon cycling, climate change is predicted to have a profound effect on river hydrology and consequently carbon export (Arnell and Gosling 2016). Flood events will become more likely, they are expected to be more severe, and increasing winter temperatures and precipitation will alter the annual cycle of river discharge, especially in boreal areas (Sonnenborg 2015). As a result of changing seasonality and magnitude of river discharge, the riverine transport of organic carbon from the catchment to the coastal zone is predicted to change as well (Whitehead *et al* 2009). In addition to changes in hydrology, increasing CO₂ concentrations and temperature have profound effects on organic carbon cycling. Elevated CO₂ concentrations have the potential to increase organic carbon production by primary producers, which in turn might lead to increased OC inputs from terrestrial to aquatic systems (Schlesinger and Andrews 2000). However, simultaneous increases in global temperatures may enhance respiration of heterotrophic microbes, thus counteracting the accumulation of soil OC (Davidson and Janssens 2006).

Changes in atmospheric deposition chemistry and catchment acid-sensitivity have also been identified as drivers behind rising OC concentrations, either coupled with climatic factors (Evans *et al* 2005, Burns *et al* 2006) or as the only driver (Vuorenmaa *et al* 2006, Monteith *et al* 2007). During the past decades atmospheric deposition of sulphate (SO₄²⁻) has decreased considerably due to cleaner industrial processes on a global scale (Engardt *et al* 2017). As high concentration of SO₄²⁻ reduces the solubility of OC by decreasing pH and increasing ionic strength, the reduced sulphate deposition has potentially increased mobilization of soil OC (Kalbitz *et al* 2000), and consequently OC concentrations in rivers (Driscoll *et al* 2003, De Wit *et al* 2007, Erlandsson *et al* 2008). However, pH in the boreal environment is not driven solely by acid

deposition, as in many peatland catchments increased organic inputs are increasing acidity (Mattsson *et al* 2007). Also, in catchments with acidic sulphate soils exposed by land uplift, acid deposition has negligible effect on pH.

Land use is known to affect specific export of OC from catchments (Wallage *et al* 2006, Clutterbuck and Yallop 2010). In Finland the highest OC concentrations and area-specific export are observed in peat-dominated catchments (Räike *et al* 2016). One third of the Finnish land area is covered by peatlands, half of which have been ditched. This has fundamental impacts on catchment hydrochemistry and hydrological processes (Holden *et al* 2004). Recently, Nieminen *et al* (2017) concluded that the ditched peatlands may contribute 2–5 times higher nitrogen and phosphorus loads to watercourses than has been previously estimated. Since nitrogen and carbon cycles are closely coupled in boreal catchments, it is also justified to expect that OC loads from drained peatlands would also be higher than current estimates.

In this study, we aimed to quantify the significance of the various drivers proposed in previous studies: climate change (increased temperature, changes in hydrology), decreased acidic deposition, and land use change by using a monitoring dataset of more than 10 000 total organic carbon (TOC) observations from 30 rivers spanning 25 years. We quantified decadal trends in TOC concentrations in Finnish boreal rivers and our objective was to determine the relative importance of different environmental factors affecting organic carbon transport from catchments to the sea. We tested this effect for each of the major drivers individually as well as their combinations. Since there is considerable variation in the properties of boreal catchments, we aimed to identify major catchment characteristics regulating the potential changes in TOC concentrations.

Material and methods

Study area

We analyzed long-term water quality monitoring data from 30 Finnish rivers, draining catchments with diverse land uses and areas ranging from 357 to 61 466 km² (figure S1 is available online at stacks.iop.org/ERL/14/124018/mmedia). All rivers discharge into the Baltic Sea with the exception of the Vuoksi River in southeastern Finland that drains into Lake Ladoga in Russia. Land use of each catchment was derived from satellite image-based land cover and forest classification data (CORINE 2006, 25 × 25 m grids). The catchments have variable hydrological and geological features, land-use patterns and population density. Due to the boreal climate and flat topography, peatlands cover one third of the total land area, half of which has been ditched for forestry. Population and industrial activities as well as agricultural activities are

concentrated in the southern and western parts of the country. Clay and silt soils are typical in the southern and western part of the country, while ground moraine soils dominate in the eastern and northern regions (Lahermo *et al* 1996).

Monitoring stations were located close to the river mouth, integrating discharges from almost the entire catchment, except for the Vuoksi River where the monitoring station was located at the Finnish-Russian border. Detailed descriptions of the sampling stations and study catchments are presented in Räike *et al* (2012). Water samples have been routinely collected at the monitoring stations, approximately with monthly frequency (table S1), and the samples have been analyzed for a broad range of substances. Sampling and analyzes were conducted within the Finnish national monitoring program maintained by the Finnish Environment Institute (SYKE).

We focused on total organic carbon (TOC), pH and water temperature, but we also used other monitoring variables for data quality assurance and in support of our findings. We used riverine pH as a proxy for changes in catchment chemistry, integrating e.g. acid deposition, liming and sulphate soil effects. Although river monitoring began in 1975, we constrained our analysis to the most recent years (1993–2017) when all 30 catchments were consistently monitored and the same analytical method for TOC was employed, providing us with a coherent and robust dataset. TOC for all observations was determined by infrared spectrometry after oxidization of the sample to carbon dioxide by combustion (according to the national standard protocol SFSEN 1484). Water temperature and pH were measured at time of sampling, using a thermometer and calibrated electrode. Our analysis included 10 964 water samples. Furthermore, daily river discharges were measured at all monitoring stations. For regulated rivers (e.g. Kemijoki, Kokemäenjoki and Kymijoki), discharge data is based on the measurements made by the hydroelectric power plants. In unregulated systems, discharge is calculated from area of water in a channel cross section and the water velocity, using discharge rating curves which describe the relationship between water level and flow in natural streams (Korhonen 2019). The water level used for flow calculations is observed daily with continuously logging pressure transducers or with chart recorders.

Statistical analyzes

Variations in TOC, pH and water temperature (T) were partitioned into seasonal and annual (calendar year) means using a decomposition method based on a general linear model (details in Carstensen *et al* 2006). This approach is well suited for heterogeneously monitoring data, as the marginal means from the general linear model account for variations in other factors of the model. TOC concentrations were log-

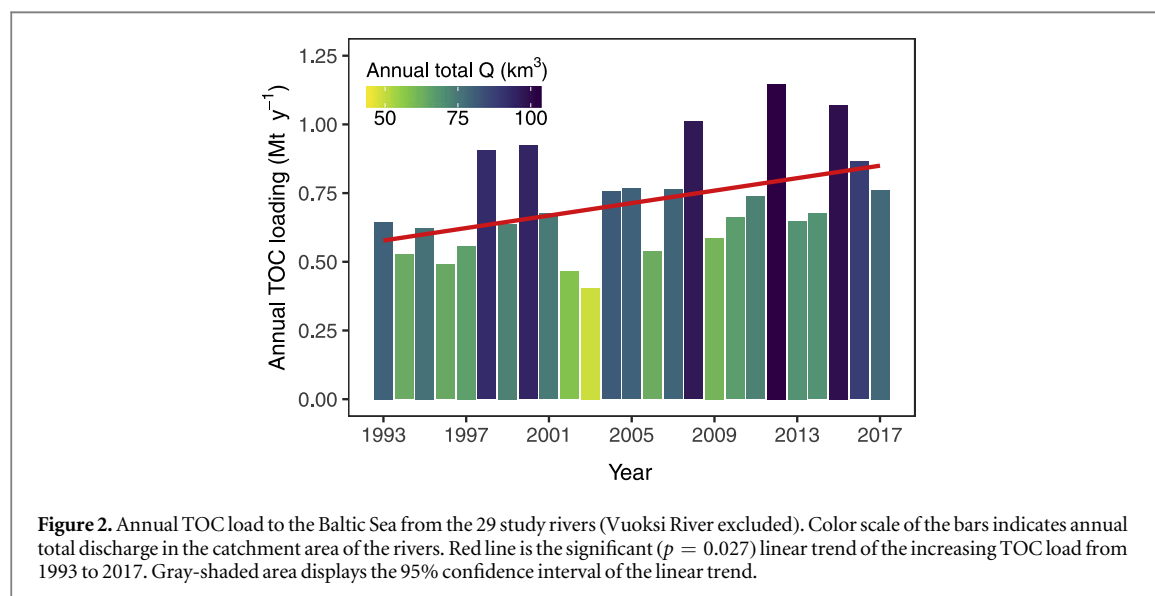
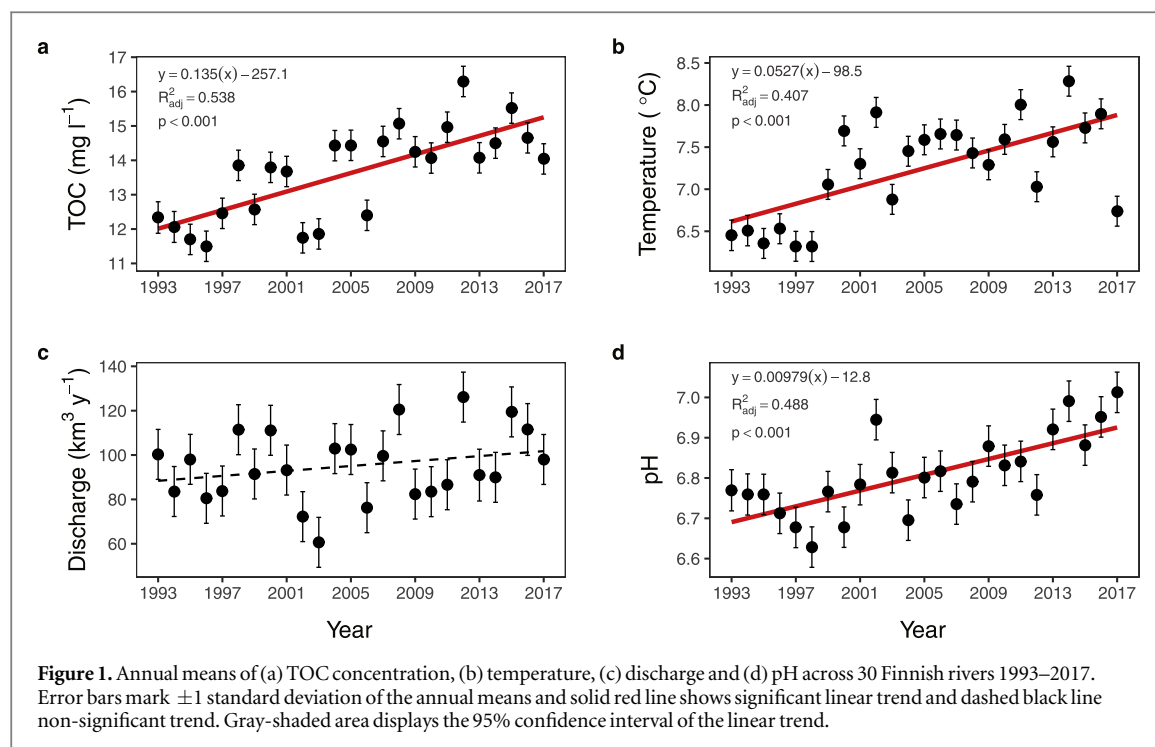
transformed prior to the decomposition to normalize distributions and the decomposition annual means were back-transformed as geometric means using the exponential function. Annual flow (Q) was calculated by aggregating daily discharges, with at least 360 observations within a single year. Annual means for all 30 catchments combined were derived from a two-way ANOVA, using year and river as categorical factors, applied to the river-specific annual means. For each river, annual means of TOC, pH and T derived from the decomposition method were combined with the annual values of Q .

Trends in TOC, pH, T and Q were examined by linear regression of river-specific annual means as well as annual means across all catchments. Residuals were examined for normality and confirmed the use of linear regression trend analysis. TOC load was calculated by multiplying the decomposed annual means of TOC concentration with respective annual mean values of discharge for each river separately, and then cumulative load of each river was aggregated as the total annual load to the Baltic Sea. The Vuoksi River was excluded from this analysis as it drains to Lake Ladoga instead of the Baltic Sea.

We analyzed potential drivers of inter-annual variations in TOC concentrations by multiple linear regression (MLR), where Q represented hydrology as driver, T represented warming as driver, and pH represented acidification as driver. MLR was employed to annual means for each river separately and regression coefficients expressed the strengths of each driver across catchments. Multicollinearity in MLR can produce strong bias among regression coefficients, but the variance inflation factor did not exceed 3 for any of the 30 regression analyzes, confirming that MLR produced reliable and relatively uncorrelated regression coefficients. Regression coefficients for the three drivers were analyzed in relation to land use in the catchment using generalized additive models (GAM). Statistical analyzes were carried out in SAS version 9.3 using the GLM, REG and GAM procedures.

Results

Our results showed an overall increase over time in TOC concentration in the 30 study rivers (figure 1(a)). On average, TOC concentration increased from 12.0 to 15.1 mg l⁻¹ between 1993 and 2017. Temperature showed significant increase as well, from 6.6 to 7.9 °C on average, although with a characteristic step-wise increase in 1999 (figure 1(b)). Annual discharge increased from 88.4 to 101.8 km³ y⁻¹, but this linear trend was not significant ($p = 0.216$). Also pH showed a significant increase, on average from 6.7 to 6.9. There was large variation among rivers, as some rivers exhibited no significant trends in TOC concentration, temperature, discharge or pH, and some showed major increases (table S2). For instance, increases in

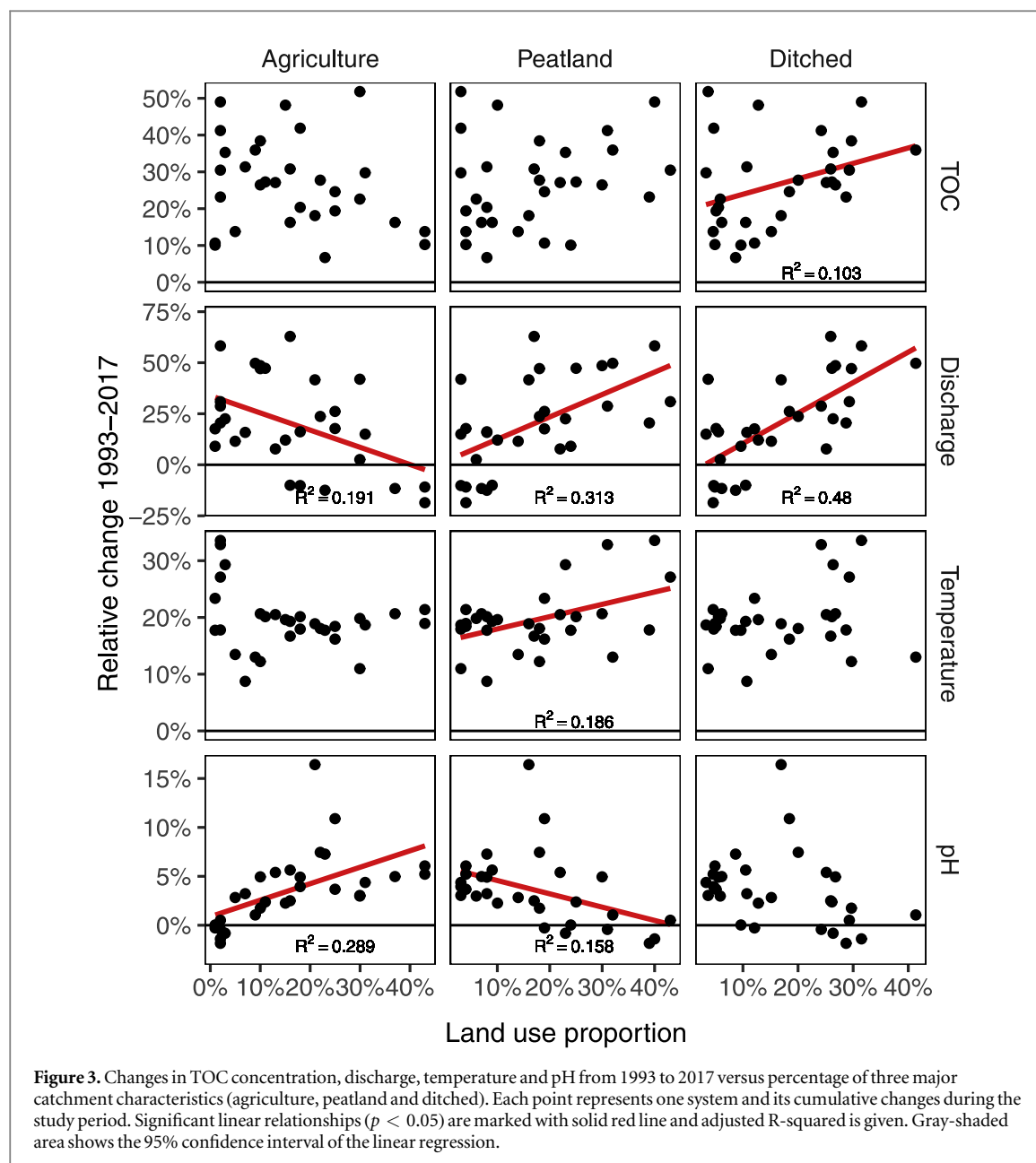


TOC concentrations from 1993 to 2017 ranged from 0.72 mg l^{-1} in the Vuoksi River to 4.54 mg l^{-1} in the Siikajoki River (table S2). Out of the 30 study rivers, only five showed no significant TOC trend (figure S2). None of the 30 rivers displayed significant negative trends for any of the study variables (figures S2–S5).

Aggregated riverine TOC load to the Baltic Sea ranged from 0.40 to 1.15 million tons per year (figure 2). TOC load showed a significant ($p = 0.027$) linear upward trend of 0.011 Mt y^{-1} during 1993–2017. During this period, freshwater discharge to the Baltic Sea from the study rivers ranged from 48.7 to $101.7 \text{ km}^3 \text{ y}^{-1}$.

The observed rates of changes in TOC concentration, discharge, temperature and pH were related to

catchment properties to a varying degree (figure 3). Discharge and pH showed opposite relationships with agriculture and peatlands, as high proportion of agriculture was linked with downward trends in discharge and upward trends in pH. In contrast, high proportion of peatlands was related to upward trends in discharge and downward trends in pH. Thus, catchments with high proportion of peatland were more susceptible to larger changes in freshwater discharge over time, whereas catchments with high proportion of agriculture were more conducive to larger changes in pH. Proportion of peatland was also correlated with temperature change, as a result of the strong north–south gradient in peatland proportion (figure S6). Proportion of ditched land in the catchment was found to

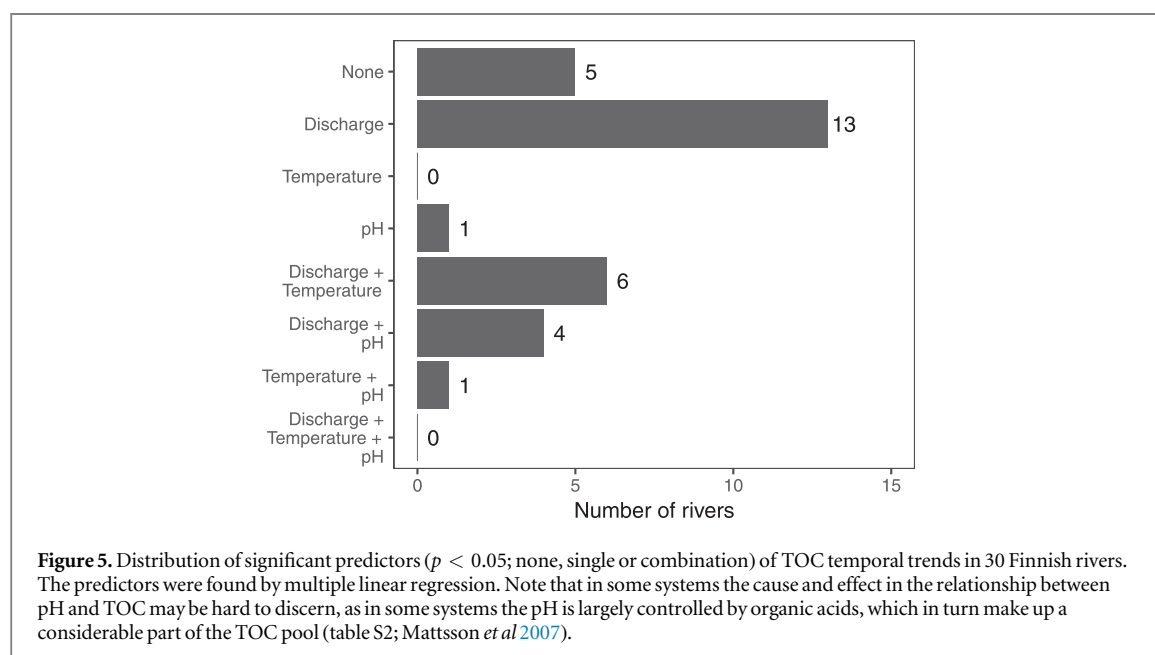
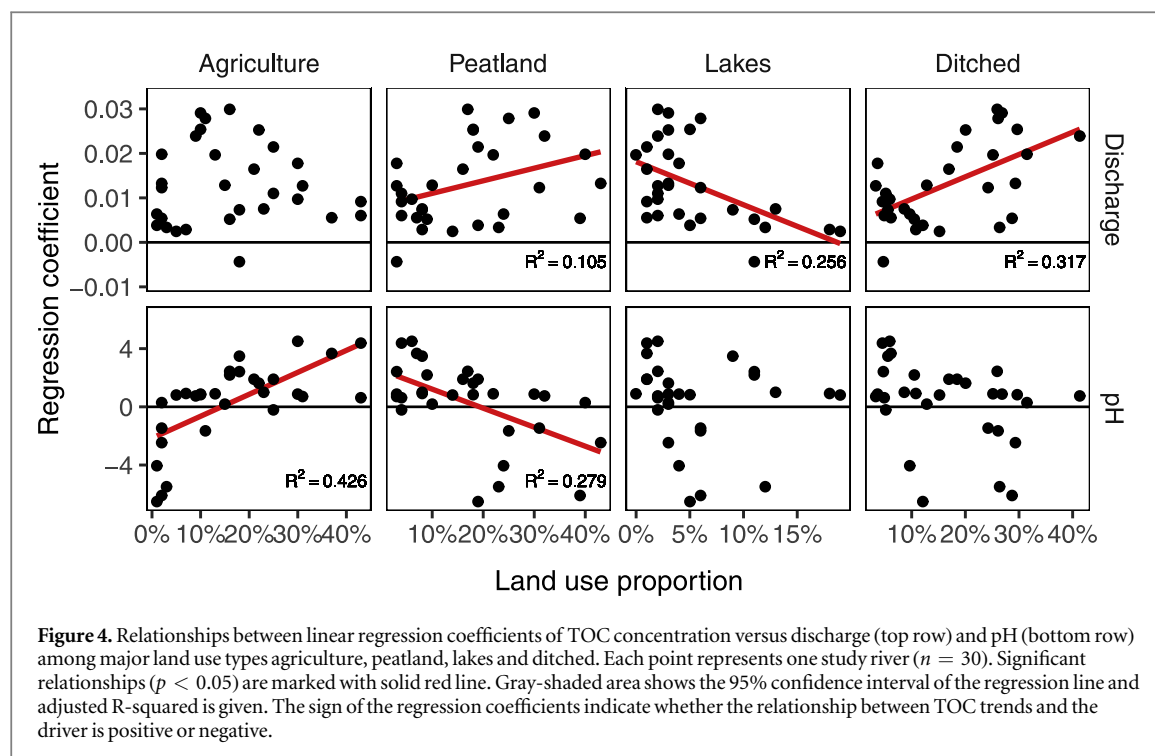


increase the relative change in both TOC concentration and discharge.

Linear regression coefficients express the strength of each driver on observed TOC concentrations across catchments. Regression coefficients of discharge had a significant negative linear relationship with lake area (figure 4), indicating that discharge was a stronger driver of changes in TOC when the lake area was small. Regression coefficients of pH were positively correlated with agricultural area of the catchment and negatively correlated with the combined forest and peatland area, indicating that pH was a stronger driver of TOC changes over time in agricultural catchments as opposed to catchments dominated by forests and peatland. The regression line of the pH coefficient versus agricultural area intercepts 0 at agricultural land use of around 20%, meaning that in catchments with low agriculture land use (less than ~20%), an increase

in pH will lead to decreases in TOC. Regression coefficients of discharge correlated positively with ditched area, meaning that in catchments with high proportion of ditched area increases in discharge led to increases in TOC concentration. Regression coefficients of temperature did not show significant relationship with any land use type (data not shown). Regression coefficients ranged from -0.004 to 0.030 , -0.15 to 1.44 and -6.51 to 4.50 with discharge, temperature and pH, respectively.

Drivers behind observed upward trends in TOC were variable among catchments. Based on the results of multiple linear regression, in 13 systems out of 25 where TOC was increasing, discharge was the most likely driver (figure 5). Discharge was also a co-predictor with other variables in ten rivers. The effect of pH was less important, as it was the single predictor in only one system and co-predictor in five systems. The

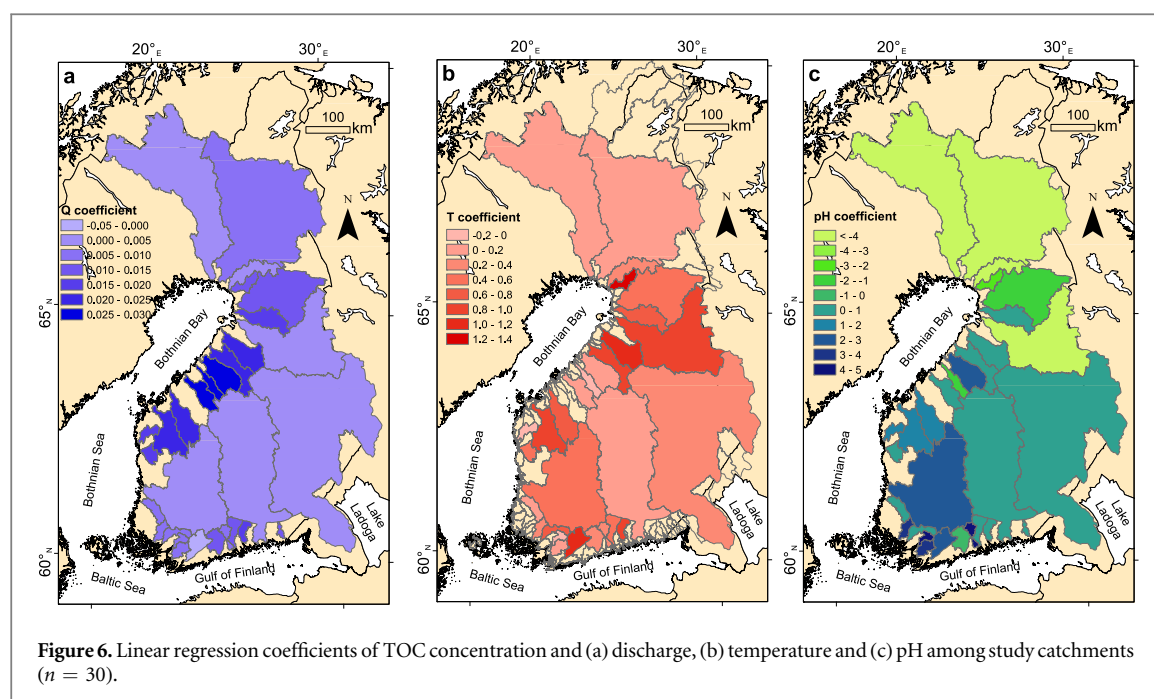


potential role of temperature driving the observed changes in TOC was significant in seven systems, but only as a co-predictor with discharge or pH.

Overall, there were no apparent geographical patterns in the spatial distribution of regression coefficients for discharge and temperature among the 30 study catchments (figures 6(a)–(b)). However, the highest regression coefficients for discharge were found in the catchments discharging to the Bothnian Bay (figure 6(a)). For pH, there was a more pronounced geographical gradient, as positive regression coefficients were typically observed in southern catchments, and negative coefficients in the north (figure 6(c)).

Discussion

We observed consistently increasing organic carbon concentrations during the past 25 years, by an annual increase of $0.04\text{--}0.25\text{ mg C l}^{-1}\text{ y}^{-1}$, varying across systems. Our results concur with the numerous upward TOC trends reported from other systems around the northern hemisphere, where annual increases up to $0.93\text{ mg C l}^{-1}\text{ y}^{-1}$ have been reported (see review by Filella and Rodríguez-Murillo 2014). The observed upward trend in TOC concentration resulted in increased TOC load to the Baltic Sea with an estimated increase of 0.28 million tons from 1993



level to 2017 level (47.2%; figure 2). Years with high TOC load coincide expectedly with high annual discharge, underlining the significance of hydrological controls to TOC loads. We also observed significant overall upward trends in temperature and pH, but not in discharge (figures 1(b)–(d)). As the upward trends were observed in multiple river systems covering extensive geographical and environmental gradients, individual driver behind observed trends may be defined.

Climate change is contributing to the observed increases in riverine TOC. Both increased precipitation and increased temperature have previously been identified as causes for TOC enrichment (e.g. Worrall *et al* 2004, Couture *et al* 2012). Increased amounts and changed temporal dynamics of precipitation have changed the spatial flow patterns and increased the surface flow in the boreal landscape, which has enhanced mobilization of organic carbon in surface soils to freshwater systems (Hongve *et al* 2004). We observed significant trends in freshwater discharge in only six rivers due to the large inter-annual variability, but overall discharge was the most important predictor for TOC changes over time (figure 4). Variation in discharge explained TOC trends in more than half of the systems, and in 13 out of 25 rivers discharge was the only significant predictor of increasing TOC. Furthermore, discharge was a significant co-predictor in five additional systems, making it evidently the most important driver. Beside trends in annual discharge, changes in the seasonality of flow have been evident in Finland and the strongest increase in TOC concentrations occurred during the winter months (Räike *et al* 2016).

Higher temperatures in the catchment may increase TOC river concentrations by either enabling

higher C fixation during longer growth season (Weyhenmeyer and Karlsson 2009) or increasing microbial mobilization of soil C (Christ and David 1996). In the former mechanism, the soil C stock stays unchanged as the increase in fixed C is primarily directed to aquatic system, whereas in the latter mechanism the soil C stock is being depleted over time. Despite being a major factor, our results show that climate change is not the only anthropogenic pressure behind increasing TOC.

Decreased anthropogenic acid depositions have increased soil pH in boreal areas, leading to increased mobilization of organic carbon (De Wit *et al* 2007). Atmospheric emission control, e.g. targeting combustion of sulphur-containing fossil fuels, has reduced the acid deposition (so-called acid rain) during recent decades. As a result, higher soil pH and changed ionic strength has increased the solubility of soil organic carbon and hence, its delivery to aquatic systems (Evans *et al* 2006). We used pH in river water as a proxy of acidic deposition. Upward pH trends were observed in 18 systems (figure S5), and pH was identified as a significant predictor of TOC trends in six systems (figure 5). However, the effect of pH is likely to become less important in coming years, as decreases in acid deposition have levelled off, stressing the importance of climate forcing even further (Erlandsson *et al* 2008). The combination of discharge and pH as main drivers of TOC was observed in multiple rivers, which could indicate some synergy in the underlying mechanism. It can be speculated, that in many catchments a change in pH is required to destabilize soil organic carbon, which will ultimately be transported to aquatic systems by the increase in surface flow. There was also a clear north–south gradient in the observations, since in the northern catchments regression coefficients

were predominantly negative, whereas in southern catchments the coefficients were mostly positive (figure 6). This gradient likely reflects the magnitude of historical acid deposition, which is the lowest in the northern parts of Finland (Engardt *et al* 2017). In five rivers, no significant driver behind observed changes in TOC was found (figure 5). The reason for this is uncertain. All of these rivers showed lower than average annual increases in TOC ($<0.136 \text{ mg l}^{-1} \text{ y}^{-1}$), and in one of the systems the increase was not significant (Paimionjoki). It is possible that the weaker trend and smaller annual variability in TOC makes it more difficult to identify the underlying drivers of change. Overall, as our data show significant contribution of three different environmental factors, it is apparent that there is no single driver explaining the increase in TOC alone, rather there is large spatial variation.

Although our study rivers covered an extensive spatial gradient displaying differing land use patterns and catchment characteristics, we observed similar trends in TOC and temperature across these broadly changing land use characteristics (figure 3). This suggests that land use does not directly intensify or lessen trends in TOC, with the notable exception of ditching: the higher the proportion of ditched area in the catchment, the steeper the increase in TOC concentrations over the 25 year study period. Previously, ditching has been shown to substantially affect stream water nutrient concentrations (Nieminen *et al* 2017). In this study, we could link, for the first time, ditching to upward trends of TOC concentrations at the downstream stations of larger rivers. This is in accordance with a recent study by R  ike *et al* (2019), linking increased nitrogen export from northern Finnish rivers to the area of ditched land.

Trends in discharge increased with the proportion of catchment peatland area, and decreased with the proportion of agricultural and urban area. Wetlands and forests attenuate the seasonal variability in stream flow compared to well-drained agricultural and urban areas, and the inter-annual component in the discharge variability might be emphasized over the seasonal component (Peralta-Tapia *et al* 2015). Land use was also linked with changes in pH, as catchments with high proportion of agriculture and low proportion of forests and peatlands showed highest changes in pH. This is likely based on the geographical distribution of agriculturally intensive areas in the south, where high depositions of sulphate in the 1980s plummeted over the following decades, while river catchments further north experienced low sulphate deposition levels throughout the study period (Vuorenmaa *et al* 2001, Helliwell *et al* 2014). Due to complex interactions of climate, geography and human activities, same environmental variable may show different temporal dynamics from catchment to another.

Catchment characteristics regulate the extent of regional or global environmental changes causing the upward trends of riverine organic carbon. Results

from the multiple linear regression analysis show that the environmental drivers change with the catchment characteristics (figure 4). Land use had no relationship with the regression coefficient for temperature, indicating that the effect of global warming on increasing TOC is independent of catchment characteristics. The effect of discharge on TOC trends was negatively related to lake area, implying that TOC in catchments with the low lake area were more sensitive to changes in discharge. These catchments lack the buffering effect that lakes in the catchment provide (Mattsson *et al* 2005).

Increasing proportion of agriculture and decreasing proportion of peatlands promoted pH as a driver of TOC variations (figure 4). In fact, TOC increased with increasing pH in catchments with large proportion of agriculture, whereas TOC decreased with increasing pH in catchments dominated by peatlands. Boreal wetlands typically have a low soil pH, compared to farmland which is more commonly mineral soil and where liming is widely used practice. A factor is also the soil type, which is in general more minerogenic in agricultural areas, and affects the organic matter characteristics transported from soils to aquatic systems (Autio *et al* 2016). The effect of different environmental drivers behind upward TOC trends can thus only be partly explained by catchment characteristics.

Increasing TOC inputs from land to coastal seas will change the functioning of coastal ecosystems. We observed an average increase from 12.0 to 15.1 mg l^{-1} in TOC concentrations between 1993 and 2017, which is equivalent to a 32% increase. As the average discharge during this period has stayed the same or even increased, this has resulted in increasing inputs of terrestrial organic carbon to coastal environment (47.2% increase from 1993 to 2017; figure 2). However, this increase has not been as pronounced or ubiquitous at Finnish coastal monitoring stations (Fleming-Lehtinen *et al* 2015). This discrepancy between TOC export from terrestrial system and concentrations in coastal waters may be explained by the functioning of the so-called coastal filter (Bouwman *et al* 2013, Asmala *et al* 2017), where different biogeochemical processes transform, retain and remove nutrients and organic matter in the coastal zone. Processes such as flocculation (Asmala *et al* 2014), photochemical degradation (Aarnos *et al* 2012) and heterotrophic microbial consumption (Fransner *et al* 2019) will reduce the amount of riverine organic carbon reaching the open sea. However, continuing increase of terrestrial inputs of organic carbon will put more pressure on already decreased environmental status of coastal areas.

In conclusion, we found significant upward trends in organic carbon concentrations in Finnish boreal rivers. The observed increases varied 6-fold among systems, and some of the systems did not show increases at all. We established that multiple environmental drivers are behind these increases, opposed to a single, ubiquitous factor. However, for some rivers, the underlying driver or drivers behind upward TOC

trends could not be established. Importantly, we could link ditching to upward trends of TOC concentrations at the downstream stations of larger rivers. Catchment characteristics can perpetuate or attenuate the effect on TOC of environmental drivers such as increases in temperature and precipitation or changes in acid deposition. Knowing the magnitude and underlying causes of increasing organic carbon trends is pertinent in understanding the loss of soil organic carbon on the one hand, and on the other hand the changes in coastal ecosystems caused by increases inputs of organic carbon.

Acknowledgments

We would like to thank David N Thomas and Hermanni Kaartokallio for insightful discussions. We are grateful to Walter and Andrée de Nottbeck Foundation for funding a workshop at Tvärminne Zoological Station. EA and JC were supported by the BONUS COCOA project (grant agreement 2112932-1), funded jointly by the EU and Danish Research Council. EA was supported by the Academy of Finland (Grant No. 309748).

Data availability statement

The data that support the findings of this study are available from the open data repository of Finnish Environment Institute (<https://syke.fi/avointieto>).

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